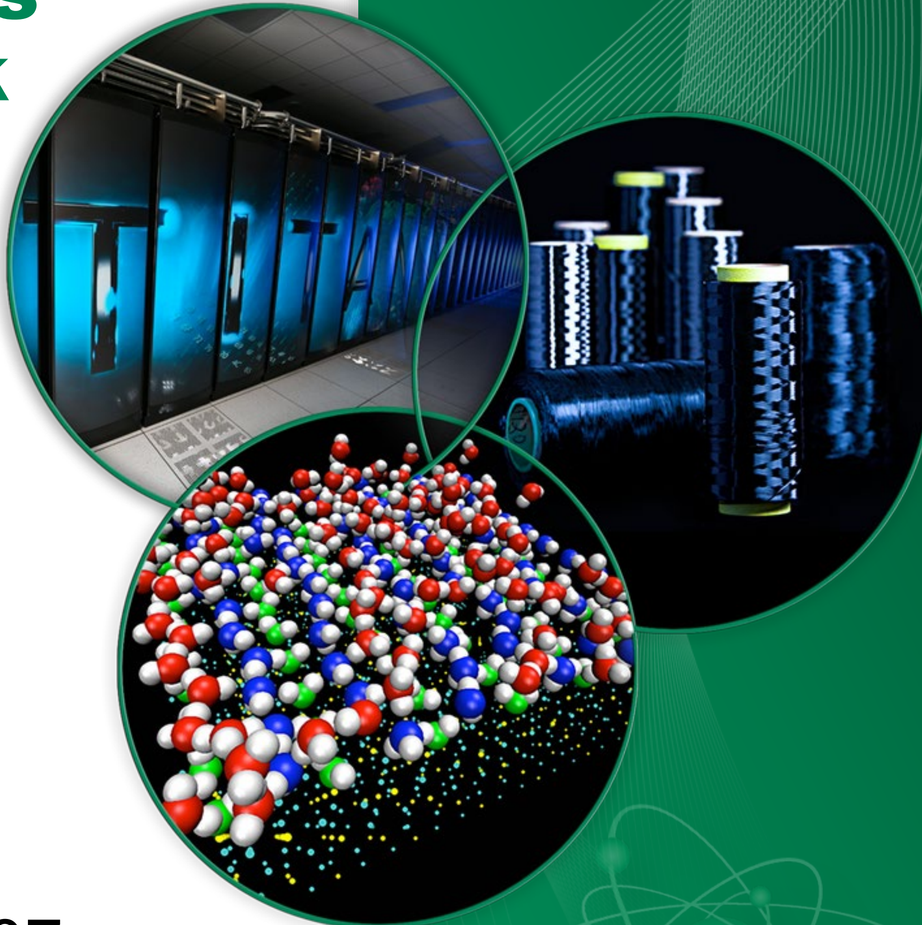
Towards Solventless Processing of Thick Electron-Beam (EB) Cured Lithium-Ion Battery Cathodes



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Oak Ridge National Laboratory

June 1-4, 2020

BAT207

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Overview

Timeline

- Task Start: 10/1/14
- Task End: 9/30/21
- Percent Complete: 70%

Barriers

- Barriers addressed
  - By 2022, further reduce EV battery cost to \$80-100/kWh.
  - Materials processing cost reduction of at least 50% and electrode thickness increase of ≥2×.
  - Achieve deep discharge cycling target of 1000 cycles for EVs (2022) at high power density.

Budget

- \$350k in FY19
- \$400k in FY20

Partners

- Interactions/Collaborations
  - Equipment Suppliers: PCT Ebeam and Integration, Keyland Polymer.
  - Battery Manufacturers: XALT Energy, Navitas Systems
  - Materials Suppliers: BASF Toda America, Allnex, Keyland Polymer.
- Project Lead: ORNL

Objectives & Relevance

- **<u>Main Objective</u>: To achieve 1) significant electrode process energy savings; 2) ultra-high electrode processing speed; and 3) utilize much more compact equipment than conventional drying ovens.**
  - EB treatment is a fast, robust materials processing technology.
  - Low cost and excellent compatibility with high-volume materials production.
  - Unmatched throughput: ≥600 m^2/min throughput can be achieved based on ≥300 m/min line speed for roll widths up to 2 m ($1.5-2.0M installed with footprint ~10 m^2).
  - Thicker electrodes: It is expected that cathode coatings of **several hundred microns** can be processed at ~150 m/min or with a larger equipment footprint.
  - Excellent energy efficiency – Electrical efficiencies ≥60% are possible.
  - Environmentally friendly – EB processing requires no solvent and no photoinitiator and has low emissions.

- **Relevance to Barriers and Targets**
  - Significantly enabling technology for achieving ultimate EV battery pack cost of $80/kWh through substantial materials processing cost reduction.
  - *Further enables cell energy density improvement through electrode thickness increases of at least 2× **<u>and</u>** is an enabling technology for solid-state battery cathodes and electrolytes.*
  - *Develops deposition methods for electrode manufacturing requiring little or no solvent.*

Task Milestones and Progress

12/31/19 (Annual Milestone Stretch): Thick electron beam (EB) cured cathode processing demonstration at ultra-high line speeds.

Demonstrate no more than 20% capacity fade through 500 cycles at 0.33C/-0.33C in 1.5 Ah pouch cells with optimized cathode EB curing formulation and NMC 811 areal loading of 30 mg/cm^2 (structured and unstructured coatings) at a curing speed of 150-200 m/min.

Completed ✓

6/30/20 (Annual Milestone Stretch): Demonstration of EB curing equipment installed at ORNL BMF.

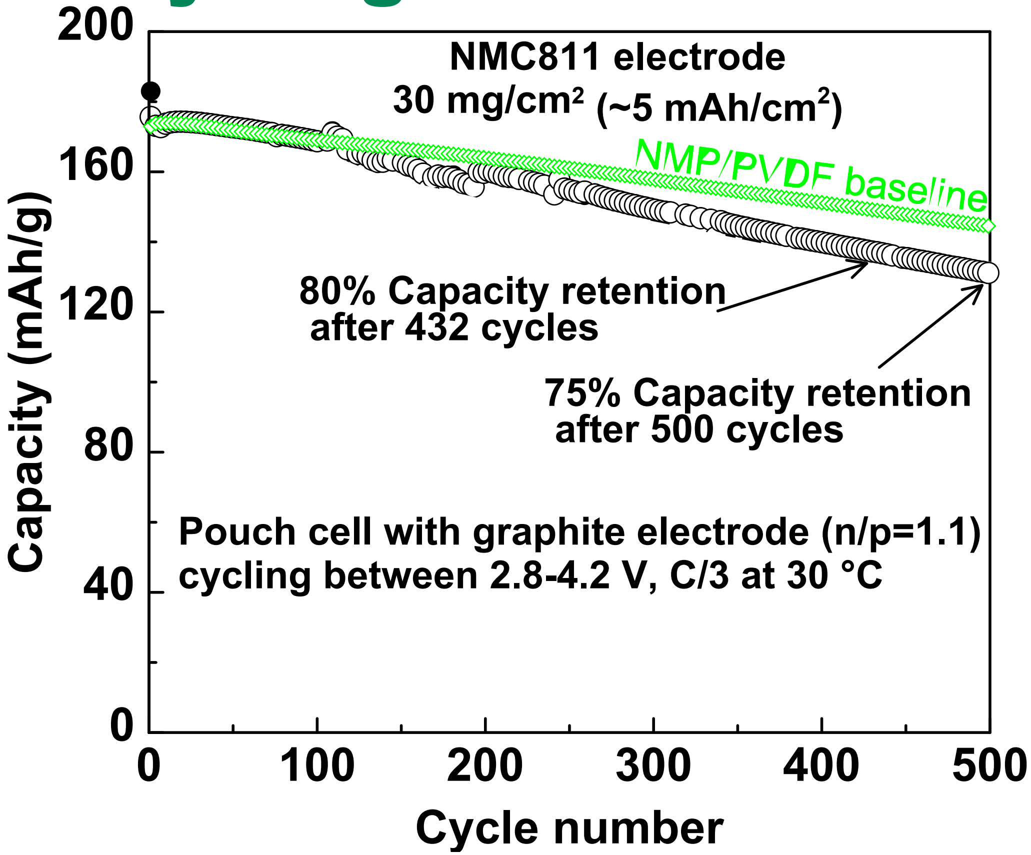
Coat a 30 mg/cm^2 NMC 811 cathode and a 25-micron PEO/LLZO composite solid electrolyte on newly commissioned PCT Ebeam R2R line at 30 ft/min.

Delayed by 3 months due to COVID-19 work restrictions; new planned completion date is 9/30/20.

Approach

- **Major problems to be addressed:**
  - Conventional solvent primary drying ovens for lithium-ion electrodes are not compatible with high line speeds or must include long drying lines to accommodate high line speed.
  - These drying lines are operating and capital expense intensive and require a large amount of battery plant space.
  - Cost of organic solvents and solvent handling are prohibitive in terms of processing cost and capital expense.
- **Overall technical approach and strategy:**
  1. Phase 1 – Demonstrate the technology's key differentiating attributes of high throughput and thick layer processing (FY15-16).
  2. Phase 2 – Address the key challenges of EB curing parameters and resulting material performance; develop ultra-thick (38 mg/cm^2 NMC 622) coating methods requiring little or no solvent. (FY17-18).
  3. Phase 3 – Demonstrate ultra-thick cathode coatings and optimized curing system in conjunction with a high-speed coating line together with a key equipment partner and battery manufacturer (FY19-20).
  4. Phase 4 – Installation, commissioning and operation of a custom roll-to-roll EB curing line at BMF and demonstration of solid-state battery cathode and electrolyte production (FY20-21).

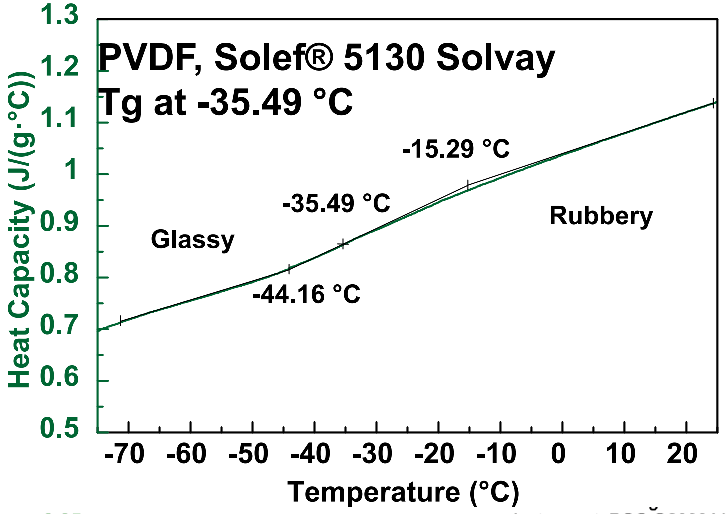
High-Speed EB Curing of Thick NMC811 Electrodes (5 mAh/cm²) Show Promising Long-Term Pouch-Cell Cycling Results

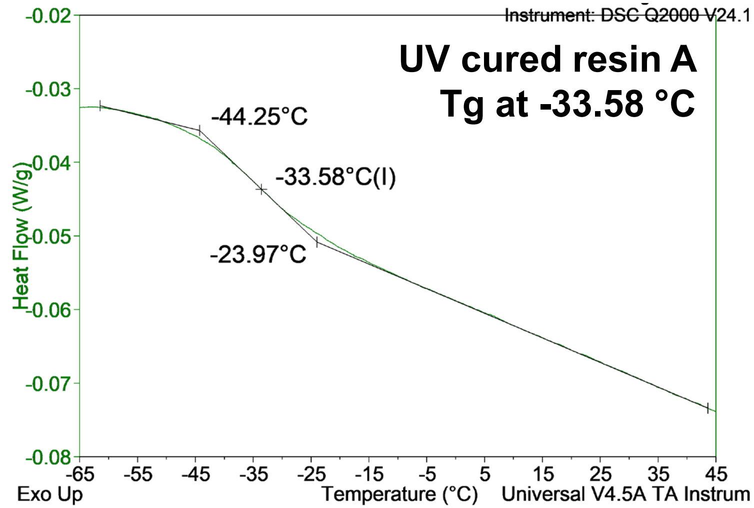


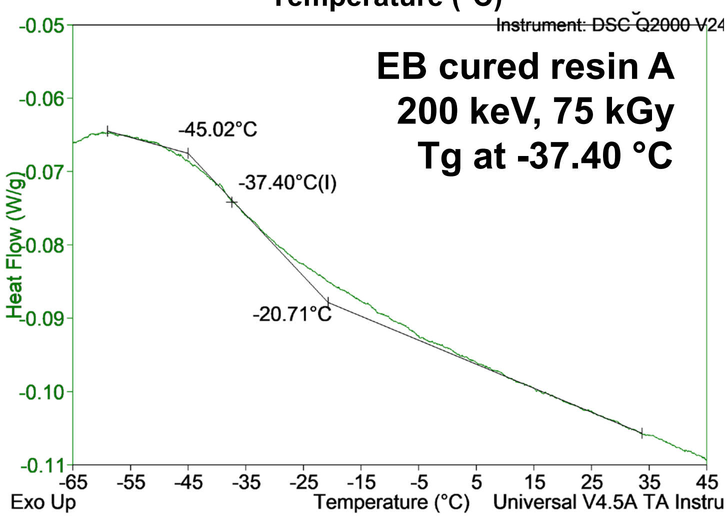
- NMC811 electrode formulation with waterborne radiation curable resins
- High speed EB Curing at PCT pilot line with line speed of 400 fpm, 75 kGy, 290 keV and 180 ppm O_2).

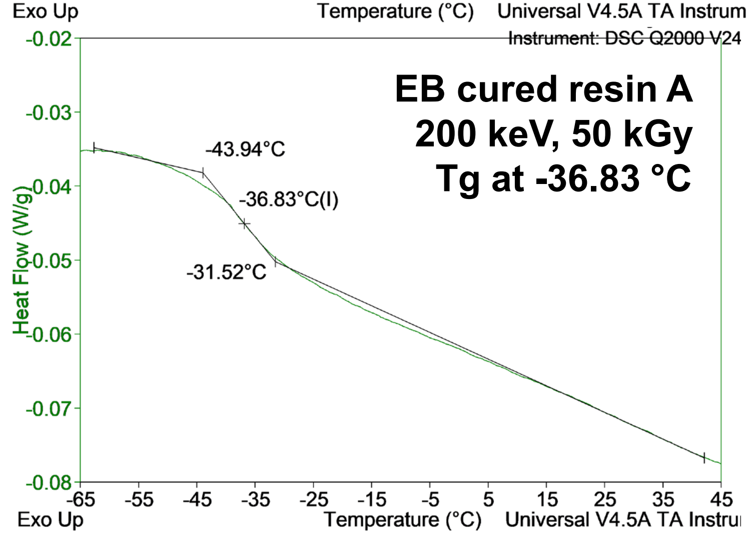
- 200 mAh pouch cells with thick NMC811 electrodes (5 mAh/cm², EB cured) show promising cycling performance.

Development of Radiation Curable Resin with Low Glass-Transition T (T_g) Similar to PVDF



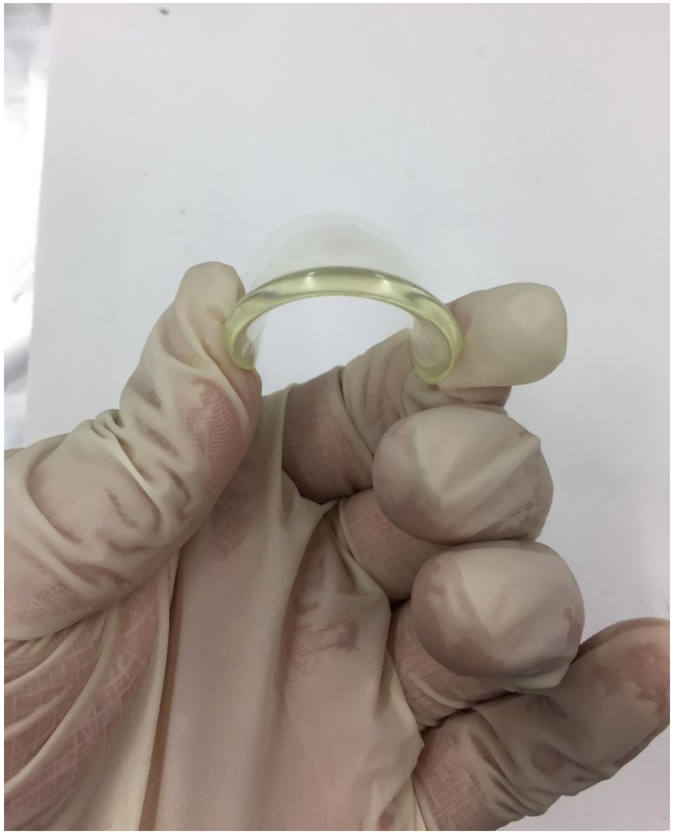




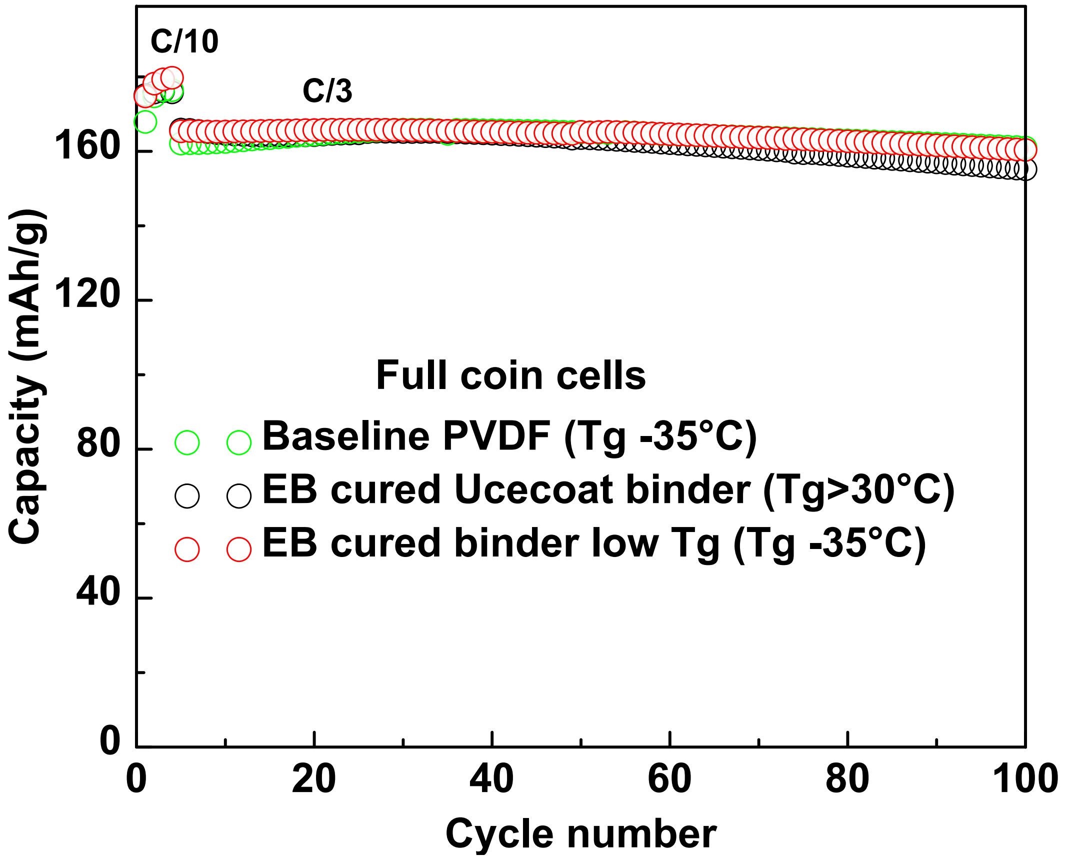


The resin is both UV and EB curable.
The cured samples have a T_g similar to that of PVDF.

Full Coin-Cell Demonstrated Good Cycling Performance Compared to PVDF Baseline

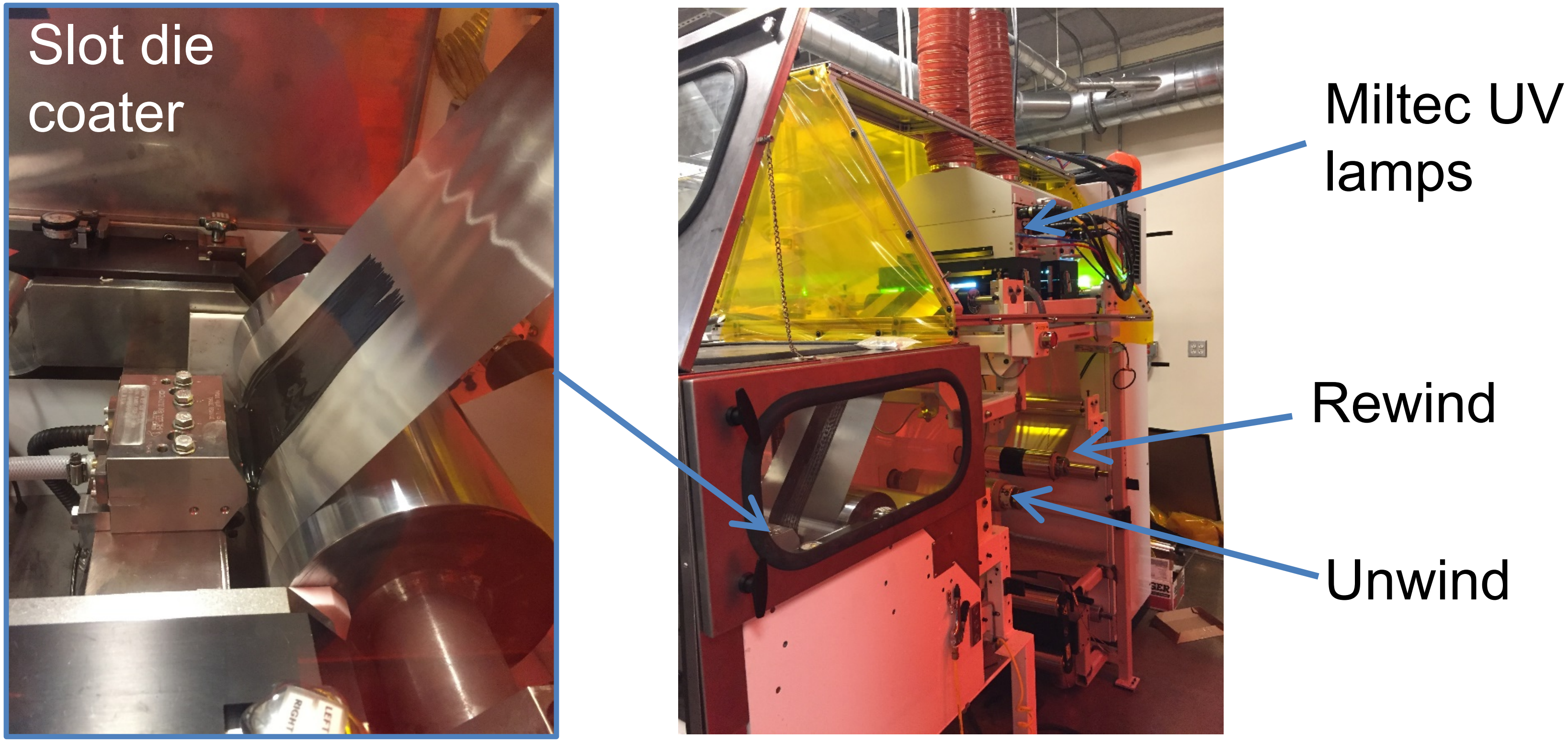


The flexibility of this new cured resin is shown here: bending of the 1 cm thick specimen.



Full coin cell testing showed little capacity fading up to 100 cycles compared to PVDF baseline and Ucecoat binders.

Implementation of R2R UV Curing Pilot Line at ORNL



- Successfully demonstrated high speed UV curing of neat resin with optimized recipe, viscosity and photoinitiator content.
- NMC composite coating has some challenges that require more optimization on electrode thickness, lamp power, line speed, etc.

New Capital Investment in Pilot Scale Roll-to-Roll EB Curing Equipment at ORNL

	Key parameters
Voltage	120-300 keV
Width	15 inches
Line speed	3-30 feet per minute
Dose	750 kGy- m/min at 300 kV
Inert	self-shielded and N_2 inerted ≤ 200 ppm of oxygen

First EB Curing of NMC Composite Electrodes Using New R2R Pilot Line

NMC electrodes with low Tg resin formulation.

Coated on Al foil using doctor blade.

EB curing under
Voltage: 300 kV
Dose: 75 kGy
O_2 level: < 200 ppm

Collaborations

- Partners
  - <u>Equipment/Coating Suppliers:</u> PCT Ebeam and Integration, NEO Beam, Keyland Polymer, B&W MEGTEC, Eastman Kodak
  - <u>Battery Manufacturers:</u> XALT Energy, Navitas Systems
  - <u>Raw Materials Suppliers:</u> TODA America, Keyland Polymer, Superior Graphite, Denka, Allnex

Selected Responses to Specific FY19 DOE AMR Reviewer Comments

The reviewer explained that the project is currently in its third phase, in which the goal is to demonstrate ultra-thick cathode coatings and an optimized curing system in conjunction with a high-speed coating line together with a key equipment partner and battery manufacturer. The reviewer asked for more information and made suggestions regarding the following: First, given the delay in delivering a piece of equipment for the roll-to-roll, EB-curing electrode-manufacturing process, the reviewer inquired how the project will be modified to guarantee timely implementation. Second, with the results demonstrated at this review meeting, it is necessary to provide electrochemical testing results, which should include multiple electrochemical techniques. For example, the reviewer wanted to know how the electrodes fabricated using the proposed processing method perform at high currents.

Thanks for the comments. As shown in slides above, the new roll-to-roll EB curing pilot line has been fabricated and is ready to use. This will accelerate the progress in this project so that more experiments can be done on site at ORNL. We have also conducted more durability tests up to 500 charge-discharge cycles to compare with baseline processing. We anticipate more electrodes available for multiple electrochemical testing techniques. High current (rate performance) will be studied.

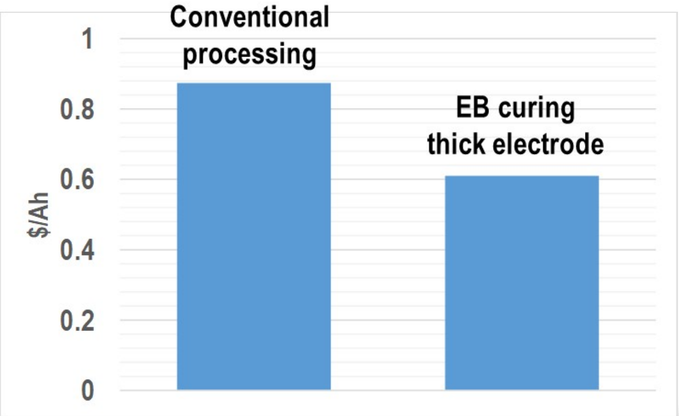
Selected Responses to Specific FY19 DOE AMR Reviewer Comments

- The reviewer noted that this project started in 2015, but progress seems to be limited by using equipment at vendors or other locations. The reviewer thought progress will accelerate when the equipment is set up at ORNL. The reviewer also noted that the pouch cell go/no-go decision in 2018 has been delayed until the third quarter of 2019, indicating the project is a bit behind.

  Thanks for the comment. As shown in the slides, we now have the equipment (both UV curing line and EB curing line) which will enable us to have more progress in 2020/2021. At the same time, we have shown 500 cycles of the pouch cells using high speed EB curing at PCT which showed promising cycling performance.

- The reviewer observed that the technical accomplishments demonstrate validity of the technique. The reviewer suggested that the techno-economic analysis should be done to see on how much this technique could reduce the overall costs of energy storage.

  Thanks for the comment. We have reported a techno-economic analysis in our FY18 slides. Based on internal cost analysis and Wood et al., J. Power Sources, 275, 234 (2015).



Proposed Future Research (FY20-21)

1. Low T_g binder formulation

   Demonstrate no more than 20% capacity fade through 500 cycles at 0.33C/-0.33C in 1.5 Ah pouch cells with optimized low T_g (less than -30°C) binder formulation and EB cured with the ORNL roll-to-roll EB curing line.

2. High solid-loading slurry formulation

   Demonstrate no more than 20% capacity fade through 500 cycles at 0.33C/-0.33C in 1.5 Ah pouch cells with optimized low T_g (less than -30°C) binder formulation and slurry solid loading (>85 wt%), and EB cured with the ORNL roll-to-roll EB curing line.

3. EB/UV R2R polymer electrolyte and composite cathode manufacturing

   Demonstrate 10 cycles of solid-state Li cell using EB/UV cured cathode and EB/UV cured polymer composite electrolyte.

Any proposed future work is subject to change based on funding levels.

Summary

- **Objective:** To achieve 1) significant process energy savings; 2) ultra-high electrode processing speed; and 3) utilize much more compact production equipment.
- **Approach:** Three-phase approach from formulation chemistry to full-scale production.
  1. Phase 1 – Demonstrate the technology's key differentiating attributes of high throughput and thick layer processing (FY15-16).
  2. Phase 2 – Address the key challenges of EB curing parameters and resulting material performance; develop coating methods requiring little or no solvent. (FY17-18).
  3. Phase 3 – Demonstrate an optimized curing system with a key equipment partner and battery manufacturer (FY19-20).
  4. Phase 4 – Installation, commissioning and operation of a custom roll-to-roll EB curing line at BMF (FY20-21)
- **Technical:** ***500 feet/min EB curing pilot line demonstration, Pouch cell performance evaluation, implementation of high-speed UV and EB curing pilot lines at ORNL.***
- **Collaborators:** High-speed EB curing scale-up at the PCT Ebeam pilot line in Davenport, IA. Plans to investigate other high-speed coating technologies with high solids (low solvent) content with either B&W MEGTEC or Eastman Kodak.
- **Commercialization:** High likelihood of technology transfer because of strong industrial collaboration, significant electrode production cost reduction, and impact on cell energy density.

Acknowledgements

- U.S. DOE Office of Energy Efficiency and Renewable Energy (EERE) Vehicle Technologies Office (Deputy Director: David Howell and Program Manager: Peter Faguy)

- ORNL Contributors:
  - Kelsey Grady
  - Andrew Todd
  - Claus Daniel

Technical Collaborators

- Michael Knoblauch
- Kevin Otto
- Karl E. Swanson
- Sage Schissel
- Michael Bielmann
- Jeff Quass
- Dave Ventola
- Kevin Dahlberg
- Lisa Stevenson
- Mike Wixom
- Dan Occorr

Information Dissemination and Commercialization

Patent & Patent Application

- A Method of Solvent-Free Manufacturing of Composite Electrodes Incorporating Radiation Curable Binders, Z. Du, C. J. Janke, J. Li, D. L. Wood, III, C. Daniel, US Patent Appl. No.: 15/966,840.
- Manufacturing of Thick Composite Electrode Using Solvent Mixtures, Z. Du, J. Li, D. L. Wood, III, C. Daniel, US Patent 10,601,027

Journal Papers and Presentations

- Z. Du, C.J. Janke, J. Li, and D. L. Wood III, “High−Speed Electron Beam Curing of Thick Electrode for High Energy Density Li-ion Batteries”, *Green Energy & Environment*, **4** (2019) 375-381.
- Kelsey Rollag, Daniel Juarez-Robles, Zhijia Du, David L. Wood, III, and Partha P. Mukherjee, *ACS Applied Energy Materials* 2019, **2** (6), 4464-4476.
- David L. Wood, III, Marissa Wood, Jianlin Li, Zhijia Du, Rose E. Ruther, Kevin A. Hays, Nitin Muralidharan, Linxiao Geng, Chengyu Mao, and Ilias Belharouak, “Perspectives on the Relationship Between Materials Chemistry and Roll-to-Roll Electrode Manufacturing for High-Energy Lithium-Ion Batteries,” *Energy Storage Materials*, Accepted.

Thank you for your attention!